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TWO LASER RAMAN DIFFERENCE TECHNIQUE APPLIED TO HIGH PRECISION SPECTROSCOPY

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We have performed precision studies of stimulated resonance Raman transitions in an atomic beam and in a vapor. The extremely narrow resonances produced by such a scheme have applications in high resolution spectroscopy and in the development of new time and frequency standards. Our studies also provide fundamental information about the nature of atom-field interaction and can be compared directly with theoretical predictions.

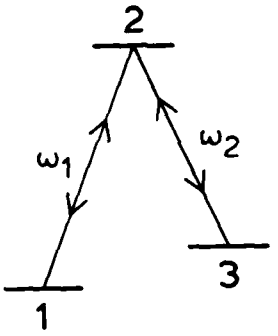

Fig. 1: *Diagram of resonance* Raman 3-level system.

Figure 1 shows schematically the stimulated resonance Raman transition under investigation. The transition is induced between two long lived states, 1 and 3, by two monochromatic laser fields at ω_1 and ω_2 which are resonant with the intermediate state 2. The total decay rate out of the i^{th} state is denoted by Γ_i. For weak copropagating pump and probe waves, the stimulated resonance Raman lineshape in a collision free Doppler-broadened system is calculated to be a Lorentzian with a width (FWHM) given by (1,2)

$$\Gamma_s = \Gamma_1 + (1 - \frac{\omega_2}{\omega_1})\ \Gamma_2 + \frac{\omega_2}{\omega_1}\ \Gamma_3 \qquad (1)$$

In a system without Doppler broadening, e.g., a highly collimated atomic beam, the expression for Γ_s reduces to

$$\Gamma_s = \Gamma_1 + \Gamma_3 \qquad (2)$$

In this paper we will present preliminary results of stimulated resonance Raman experiments that we performed in a vapor and also in an atomic beam.

Resonance Raman Studies in a Vapor

We have used the system shown in Fig. 2 to study resonance Raman transitions in I_2 vapor. The pump laser, a single mode argon laser operating at 5145 Å, is short-term stabilized by locking it to a passive Fabry-Perot reference cavity (3). A single mode fiber which couples pump laser radiation into this cavity eliminates misalignment errors due to laser beam steering and permits the isolation of the cavity from the noisy environment of the water-cooled argon laser. The frequency ω_1 of the pump laser is long term stabilized by locking it to an I_2 hyperfine resonance in a saturation spectrometer (3). The probe laser, a single mode dye laser operating at 5828 Å, is short term stabilized to another passive Fabry-Perot cavity. The long term stability of the pump frequency is transferred to the probe via an evacuated transfer Fabry-Perot cavity as shown in Fig. 2. Part of the pump laser radiation at ω_1 is shifted by an acoustooptic modulator operating at frequency δ. The shifted pump beam is coupled into the transfer cavity, and the cavity is locked to the long term stabilized frequency $\omega_1 + \delta$. The dye laser frequency is then long term stabilized by locking it to a transmission peak of the transfer cavity. In this manner the dye laser can be precisely tuned by changing the frequency δ of the RF drive to the A/O modulator.

In this experiment the argon laser pump is amplitude modulated at 2 kHz and combined with the dye laser probe. Both beams are then expanded to ~ 1 cm dia. for one pass through a 150 cm long cell containing I_2 vapor. To eliminate misalignment effects due to laser beam steering, the pump and the probe are coupled through a fiber prior to entering the cell. This second fiber also acts as a spatial filter for both beams and insures that the pump and the probe are collinear. A prism after the cell separates the pump from the probe which is synchronously demodulated as shown in Fig. 2. Intensity

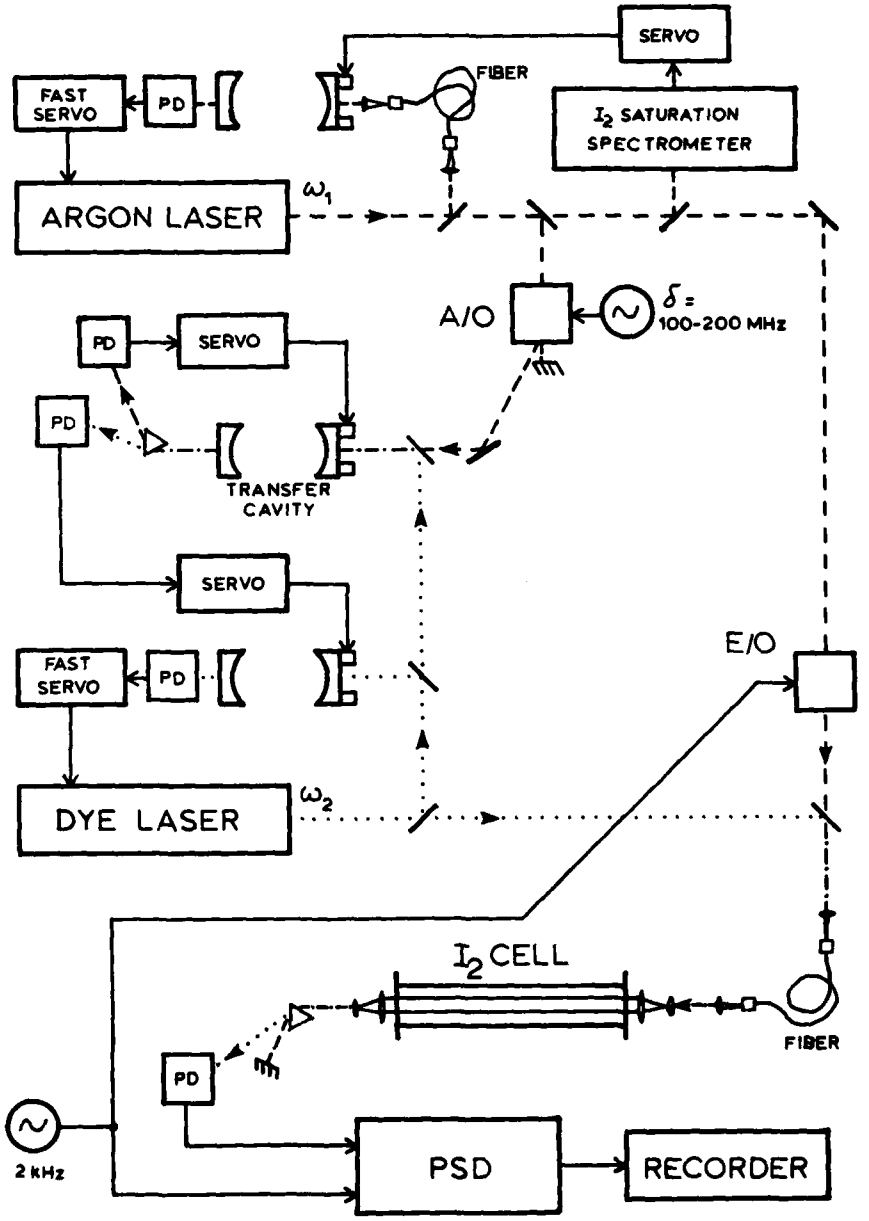

Fig. 2 Experimental set-up for I_2 cell studies.

subtraction (not shown) of probe beam fluctuations reduces background noise to less than 1.5 times the shot noise limit.

In the I_2 molecule under study, states 1 and 3 are hyperfine components of the v" = 0, J" = 15 and v" = 11, J" = 15 rovibrational levels in the ground electronic state. These states have essentially zero natural width, so Γ_1 and Γ_3 are limited by the transit time. Level 2 is one of the hyperfine components of the v' = 43, J' = 16 level of the first excited electronic state with $\Gamma_2 \sim 100$ kHz. Figure 3 shows the experimental weak field lineshape which has a width (FWHM) of 58 kHz. (I_2 pressure is less than 1 mtorr.) The experimental linewidth is presently determined by transit time effects, a residual contribution from Γ_2 and dye laser jitter.

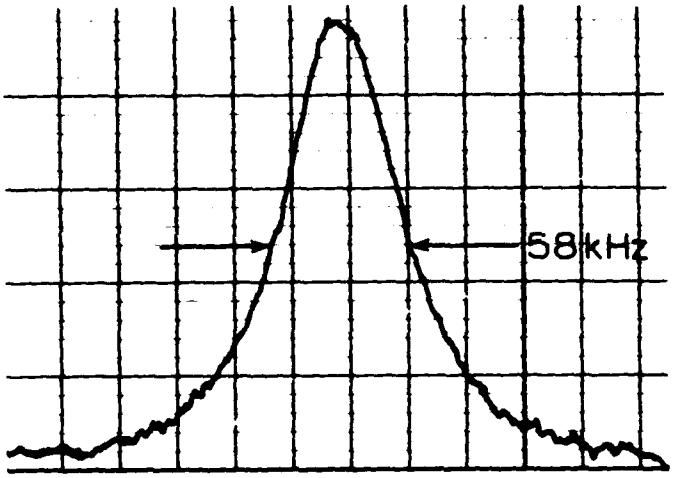

Fig. 3 Weak field resonance Raman lineshape.

The extremely narrow resonances generated by this experiment are attractive candidates for secondary frequency standards in the visible region (2). These resonances can also be used for Doppler-free stimulated emission spectroscopy of thermally unpopulated levels and as unique high resolution probes for the study of collisional effects on specific energy levels (2). For counterpropagating pump and probe (4), the width Γ_s of the Raman transition depends primarily on Γ_2. We are using this effect to study differences in the relaxation rates of individual I_2 hyperfine levels due to magnetic predissociation (5). Finally, one can use the lineshape produced by a strong pump and a weak probe to study the ac Stark effect in a Doppler-broadened system and to obtain average matrix elements for individual I_2 hyperfine transitions (6).

Resonance Raman Studies in an Atomic Beam

We have used the experimental setup illustrated in Fig. 4 to study resonance Raman transitions in a sodium atomic beam. As shown in Fig. 4, two dye laser beams at ω_1 and ω_2 are combined on a beam-splitter to produce beams A and B. The ω_1 and ω_2 components of both beams A and B are weak and copropagating. Atomic beam fluorescence induced by beam B is collected with a photomultiplier. Changes in this induced fluorescence are monitored to obtain resonance Raman lineshapes. For single region excitation, ω_1 and ω_2 are generated by two short term stabilized dye lasers similar to those described earlier.

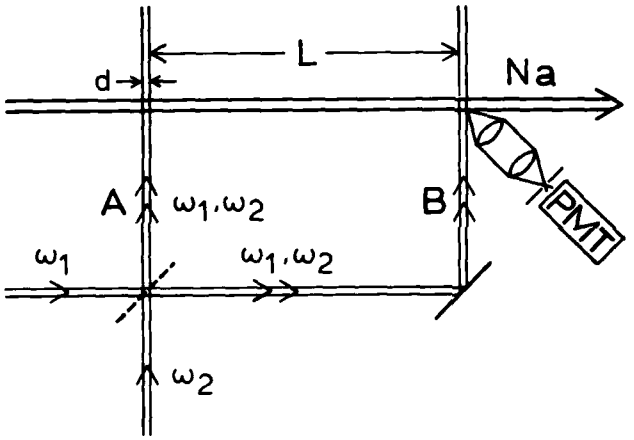

Fig. 4 Experimental set-up for Na atomic beam studies.

In this case, states 1 and 3, Fig. 1, are the $3^2S_{1/2}$(F=1) and $3^2S_{1/2}$(F=2) ground sublevels in atomic sodium, respectively, separated by 1772 MHz, and level 2 is the $3^2P_{1/2}$ (F=2) level, having a 16 ns lifetime. Data is obtained by holding ω_1 on resonance with the 1 2 transition while tuning ω_2 over the 3 2 transition frequency. To partially suppress background fluorescence and enhance signal to noise the laser beam at ω_1 is chopped and the collected beam B fluorescence is demodulated using a lock-in amplifier.

Figure 5 shows the lineshape for a stimulated resonance Raman transition in a single interaction region using zero magnetic field. The narrow, 200 kHz (FWHM) dip in the center of the trace is the Raman transition. Its width, as predicted by equation (2), is primarily determined by transit time and is independent of the width of state 2. The broad feature in the same trace is a small segment of

the 10 MHz wide spontaneous emission background from state 2.

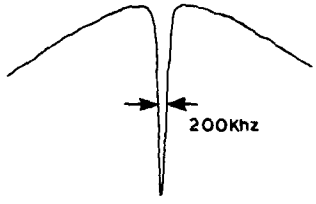

Fig. 5 Resonance Raman lineshape in Na atomic beam.

In order to reduce the transit time linewidth we use separated field excitation (7), as shown in Fig. 4, with beams A and B both allowed to interact with the atomic beam. Lineshapes obtained with this setup are commonly referred to as Ramsey fringes (8). To obtain fringe lineshapes corresponding to large interaction region separations, laser jitter in the two lasers must be well correlated (7). In our experiment, because of the small 1772 MHz separation between states 1 and 3, it is possible to do this by generating ω_2 directly from ω_1 using an acoustooptic modulator. Figure 6 shows a fringe of width 1.3 kHz (FWHM) corresponding to a 30 cm interaction

Fig. 6 Ramsey fringes using resonance Raman excitation in Na atomic beam.

region separation. For this trace, beam A is chopped, instead of the laser beam at ω_1. A small Zeeman field of approximately 300 mG is applied along the laser propagation direction to separate out the effects of transitions other than the $m_F = 0$, $\Delta m_F = 0$ Raman transition. The acoustooptic driver which determines $(\omega_1 - \omega_2)$ has a short term stability of less than 100 Hz.

We plan to investigate the applicability of stimulated, resonance Raman techniques in atomic beams to frequency standards and high resolution spectroscopy. Of particular interest is the possibility of extending this Raman technique into the millimeter and

even FIR regions of the spectrum.

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